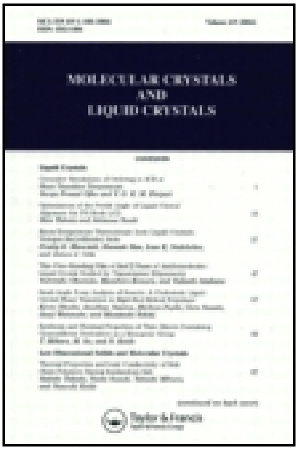

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Continuum Theory of Liquid Crystals of Nematic Type

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Abstract—We describe continuum theory now being used to describe macroscopic behavior of liquid crystals of nematic type. Two types of predictions are discussed. The first concerns the propagation of orientation waves. The second area is viscometry, including effects of simple electromagnetic fields. We discuss some unusual size effects predicted when such fields are absent.

Introduction

The continuum theory of liquid crystals to be discussed is obtained by wedding a rather successful static theory to one which seems to be consistent with viscometric measurements at higher rates of shear. It seems fair to say that any adequate theory should include, in some approximation, this one. Various types of generalizations have been mentioned by different writers. This theory is a special case of the class of theories considered by Leslie,[1] who allows for more nonlinearity. Oseen[2] sketches a theory accounting for longer range interactions. Coleman[3] and Wang[4] emphasize theories involving long memories. Peter and Peters[5] discuss the desirability of accounting for fractional orientation. Akin to this are the ideas involved in swarm theories, discussed, for example, by Gray[6] (*cf.* Ch. IV). It is not difficult to think of additional, more or less plausible types of generalization.

It is not entirely trivial to cope with the additional difficulties associated with such generalizations. It thus seems to me sensible to obtain predictions from the simpler theory and to attempt to clarify in what respects theory is inadequate to describe experiment, a matter which is not yet entirely clear.

We here limit our attention to liquid crystals of nematic type and to two areas of research; propagation of orientation waves and viscometry. Other types of predictions are mentioned in a recent survey by Ericksen.[7]

Basic Equations

For present purposes, we consider the fluids to be incompressible and ignore thermal effects. In Cartesian tensor notation, the governing equations are of the form

$$d_k d_k = 1 \tag{1}$$

$$v_{k,k} = 0 \tag{2}$$

$$\rho\, dv_k/dt = \sigma_{jk,j} + f_k \tag{3}$$

$$\rho_1\, d^2 d_k/dt^2 = \pi_{jk,j} + g_k + h_k \tag{4}$$

$$\sigma_{ji} = -p\delta_{ij} - \pi_{jk}\, d_{k,i} + \hat{\sigma}_{ji} \tag{5}$$

$$\pi_{ji} = \partial W/\partial d_{i,j} \tag{6}$$

$$g_i = \gamma\, d_i - \partial W/\partial d_i + \hat{g}_i \tag{7}$$

$$\begin{aligned} 2W = {} & k_{22}\, d_{i,j}\, d_{i,j} + (k_{11} - k_{11})(d_{k,k})^2 \\ & + (k_{33} - k_{22})\, d_i\, d_j\, d_{k,i}\, d_{k,j} + k_{24}[d_{i,j}\, d_{j,i} - (d_{k,k})^2] \end{aligned} \tag{8}$$

$$\begin{aligned} \hat{\sigma}_{ji} = {} & \mu_1\, d_k\, d_p A_{kp}\, d_i\, d_j + \mu_2\, d_j N_i + \mu_3\, d_i N_j + \mu_4 A_{ij} \\ & + \mu_5\, d_j\, d_k A_{ki} + \mu_6\, d_i\, d_k A_{jk} \end{aligned} \tag{9}$$

$$\hat{g}_i = \lambda_1 N_i + \lambda_2\, d_j A_{ji} \tag{10}$$

$$N_i = d\, d_i/dt + 1/2(v_{k,i} - v_{i,k})\, d_k \tag{11}$$

$$2A_{ij} = v_{i,k} + v_{k,i} \tag{12}$$

Here d_k is a unit vector, denoting the preferred direction inherent in liquid crystals, v_k being the velocity vector. Associated with the constraints of incompressibility and (1) are two arbitrary functions p and γ, essentially Lagrange multipliers. The k's, λ's and μ's are material constants, related as follows

$$\lambda_1 = \mu_2 - \mu_3, \qquad \lambda_2 = \mu_5 - \mu_6 \tag{13}$$

Further, ρ is the constant mass density, ρ_1 being a positive constant, another inertial coefficient. In a fluid macroscopically at rest, there can be inertia associated with molecules spinning about their centers of mass, measured by ρ_1.

The moduli should satisfy two kinds of inequalities. One, essentially a stability condition, guarantees that $W \geqslant 0$. As is discussed by Ericksen,[8] the k's should then satisfy

$$k_{11} \geqslant |k_{11} - k_{22} - k_{24}|, \qquad k_{22} \geqslant |k_{24}|, \qquad k_{33} \geqslant 0 \tag{14}$$

Other restrictions follow from the Clausius inequality, as is discussed by Leslie[1,8], *viz.*

$$\mu_4 \geqslant 0, \qquad 2\mu_1 + 3\mu_4 + 2(\mu_5 + \mu_6) \geqslant 0, \qquad 2\mu_4 + \mu_5 + \mu_6 \geqslant 0$$
$$\lambda_1 \leqslant 0, \qquad -4\lambda_1(2\mu_4 + \mu_5 + \mu_6) \geqslant (\mu_2 + \mu_3 - \lambda_2)^2 \tag{15}$$

In (3) and (4), f_k and h_k represent the effect of external actions, f_k being a body force per unit volume, $\epsilon_{klm}\, d_l h_m$ being a body couple per unit volume. The latter essentially determines h_m since any part of h_m which is parallel to d_m can be absorbed in the multiplier γ. For example, in the case of an imposed static magnetic field H_k, we might argue that this produces a magnetization M_k, given by a linear relation of the form

$$M_i = kH_i + lH_j\, d_j\, d_i \tag{16}$$

where k and l are constant susceptibilities. According to elementary estimates, this gives rise to a force

$$f_i = M_k H_{i,k} \tag{17}$$

and couple

$$\epsilon_{ijk} M_j H_k = \epsilon_{ijk}\, d_j (lH_m\, d_m H_k) \tag{18}$$

per unit volume, so we can take

$$h_k = lH_m\, d_m H_k \tag{19}$$

Such is the estimate used in interpreting measurements wherein static orientation patterns are influenced by such fields. This provides a means for obtaining experimental values of k_{11}, k_{22} and k_{23}, as is discussed by Saupe.[9] The term multiplied by k_{24} drops out of the equation of motion. It does influence surface couples, which are expressible in terms of π_{jk}. It seems that no thought has been given to means for measuring these. In (3), σ_{jk} is the stress tensor. Since (9) relates part of this to velocity gradients, we might expect to obtain values for some of the μ's from viscometric data. Alternatively noting (13), we might emphasize (10) and (4), exploring the effects of changing orientation. The latter may involve ρ_1. Said differently, we might explore orientation waves. We now turn to some of the more recent theory pertaining to these possibilities.

Orientation Waves

After J. L. Fergason described to me preliminary observations concerning the propagation of disturbances in orientation, I began to explore what analyses of this might be feasible. The simplest case would be when there is no accompanying disturbance in velocity. Theoretically, this is quite exceptional. We[10] found one case of this type, a solution of the form

$$\mathbf{d} = (\cos\theta, \sin\theta, 0), \qquad \theta = \theta(z, t), \qquad f_k = h_k = 0 \tag{20}$$

θ being any solution of the linear wave equation

$$\rho_1 \frac{\partial^2\theta}{\partial t^2} = \lambda_1 \frac{\partial\theta}{\partial t} + k_{22}\frac{\partial^2\theta}{\partial z^2} \tag{21}$$

As might be expected, inequalities (14) and (15) imply that the wave speed $\sqrt{k_{22}/\rho_1}$ is real, the damping coefficient λ_1 negative.

It is also possible to obtain some information by studying wave fronts associated with pulses. Mathematically, we study conditions which must hold on surfaces across which there are discontinuities in orientation and motion. If d_i, v_i and their first derivatives are continuous, but higher derivatives have finite discontinuities, I[11] find that the discontinuity surface S moves relative to the material with a speed U given by

$$\rho_1 U^2 = k_{22} + (k_{33} - k_{22})(d_k \nu_k)^2 \qquad (22)$$

or

$$\rho_1 U^2 = k_{11} + (k_{33} - k_{11})(d_k \nu_k)^2 \qquad (23)$$

ν_k denoting the unit normal to S. The first applies in cases where the orientation discontinuity is perpendicular to d_k and ν_k, the second applying when it is in the plane determined by d_k and ν_k, being perpendicular to d_k. The inequality (14) implies that all these speeds are real. Associated with the discontinuity in orientation, there is a calculable discontinuity in motion, which will generally promote dissipation of the wave. As yet, no other predictions have been worked out, though it would seem feasible to develop a much more complete theory. I have not seen quantitative data on wave speeds, which might be used to obtain an estimate of ρ_1 and to supplement static measurements of the k's. One can of course use simple molecular models to estimate ρ_1, but it would be comforting to have relevant data.

Size Effects in Viscometry

Statically, there is the familiar fact that a solid in contact with a liquid crystal tends to produce a certain orientation, depending on the nature and prior treatment of the solid. It is also rather clear that a shear flow promotes a certain orientation. In viscometry, there is thus the possibility of competition arising in cases where these two orientations differ. Roughly, we might expect the wall to dominate if the flow is very slow or if the gap is very narrow, the shear flow dominating if it is fast, the gaps wide. Of course, it is possible that size effects might result from entirely different causes. That size effects are observed is mentioned by Porter and Johnson.[12] Porter, Barrall and Johnson[13] suggest that the competition here mentioned may be responsible for higher viscosities observed at lower rates of shear. It thus seems pertinent to inquire what theory predicts.

Consider flow between parallel flat plates, a distance h apart, one stationary, the other moving with speed V. The nominal shear rate is then

$$\dot{\gamma} = V/h \tag{24}$$

If we now assume

(a) $f_k = h_k = 0$,

(b) the orientation at the plates is independent of $\dot{\gamma}$,

(c) the " molecular inertia " $\rho_1 d^2 d_i/dt^2$ is negligible,

Ericksen's[14] scaling argument indicates that the apparent viscosity should scale as indicated by the equation

$$\mu = f(\dot{\gamma}h^2) \tag{25}$$

consistent with the idea that decreasing $\dot{\gamma}$ is equivalent to decreasing h. For a fixed liquid crystal, the form of this function will be different for different choices of wall orientation. In an earlier analysis, Leslie[1] obtained a solution for a particular case which exemplifies (25). For it, η approaches finite limits as $\dot{\gamma}h^2 \to 0$ and as $\dot{\gamma}h^2 \to \infty$. The former corresponds to the apparent viscosity corresponding to uniform orientation at the orientation preferred by the wall. The latter limit corresponds to the apparent viscosity corresponding to uniform orientation at the orientation preferred by the flow.

Porter and Johnson[12] mention that the apparent viscosity seems abnormally high in very narrow gap instruments at higher rates of shear. Then (25) indicates that it should increase with decreasing shear rate, in a fixed instrument, which is observed. Peter and Peters[5] attribute the latter effect to fractional orientation. It is of course conceivable that boundaries might influence the degree of orientation, leading also to size effect.

It is possible to develop similar scaling rules for the various viscometers, using similar assumptions. For example, Atkin[15] shows that, for Poiseuille flow,

$$Q = rg(r^3\mathscr{P}) \tag{26}$$

where $\mathscr{P}$ is the driving pressure gradient, r the radius of the capillary and Q the flow rate.

To calculate the form of the function involved in, say (26), we would need experimental information concerning orientation at the wall, which seems not to be available. Empirical scaling rules remain to be established, so we cannot check theory in this regard. Little attention seems to have been given to the possibility that the material from which the viscometer is made might influence viscometric response. It should be noted that, to the extent that such size effects occur, it is unsound to use familiar data reductions to extract an apparent viscosity curve from raw data. In brief, it will take more effort to gain a firm understanding of the phenomena occurring in viscometry.

Some simplification results when we restrict our attention to the high rate of shear, wide gap regime, wherein the flow dominates. Formally, we set

$$k_{11} = k_{22} = k_{23} = 0 \tag{27}$$

in the governing equations, and relinquish the possibility of specifying orientation at boundaries. Experimentally, this involves some trial and error, to find the range where different instruments yield, by standard data reductions, consistent apparent viscosities. Some such correlations are discussed by Porter, Barrall and Johnson.[13] Henceforth, we restrict our attention to this regime, adopting the simplification (27).

Orientation Induced by Shear

For a simple shearing flow,

$$v = (\dot{\gamma} x_2, 0, 0), \qquad \dot{\gamma} > 0 \tag{28}$$

two quite different types of behavior are possible, according as

$$|\lambda_1/\lambda_2| \leqslant 1 \tag{29}$$

or

$$|\lambda_1/\lambda_2| > 1 \tag{30}$$

When (29) applies, **d** tends toward a steady state value, lying in the plane of shear, making an angle ϕ with the x_1-axis, where

$$\cos 2\phi = -\lambda_1/\lambda_2 \tag{31}$$

When (30) applies, **d** varies periodically with time. An analysis of this, neglecting the " molecular inertia ", is given by Ericksen.[16] Experimental indications such as are discussed by Porter and Johnson[12] suggest that (29) applies, though we are at least close to the borderline between the two conditions. That is, ϕ is close to zero, or

$$\lambda_1/\lambda_2 = -1 + \epsilon \tag{32}$$

where ϵ is a small positive number, if not zero. If **d** is not at its steady state value, but is varying with time, the stresses will vary with time, as is obvious from (9). We here disregard such relaxation effects.

For any constant value of **d**, we can compute the stresses. Generally, none of the stress components vanish. The shear stresses and normal stress differences are proportional to $\dot{\gamma}$, with factors of proportionality depending on **d**. If **d** is either in or perpendicular to the plane of shear, the only cases which we consider,

$$\begin{aligned}
\sigma_{13} = \sigma_{31} &= \sigma_{32} = \sigma_{23} = 0,\\
2\sigma_{21} &= \dot{\gamma}[2\mu_1 d_1^2 d_2^2 + \mu_4 + (\mu_5 - \mu_2) d_2^2 + (\mu_6 + \mu_3) d_1^2],\\
2\sigma_{12} &= \dot{\gamma}[2\mu_1 d_1^2 d_2^2 + \mu_4 + (\mu_5 + \mu_2) d_1^2 + (\mu_6 - \mu_3) d_2^2],\\
2(\sigma_{11} - \sigma_{33}) &= \dot{\gamma}\, d_1 d_2(2\mu_1 d_1^2 - \mu_2 - \mu_3 + \mu_5 + \mu_6),\\
2(\sigma_{22} - \sigma_{33}) &= \dot{\gamma}\, d_1 d_2(2\mu_1 d_2^2 + \mu_2 + \mu_3 + \mu_5 + \mu_6)
\end{aligned} \tag{33}$$

The usual definition of apparent viscosity is

$$\mu = \sigma_{21}/\dot{\gamma} \tag{34}$$

and, of course, **d** is restricted to be a unit vector. The inequalities (15) imply μ is positive. If (32) holds with $\epsilon = 0$, and **d** is at its steady state value

$$\mathbf{d} = (1,0,0)$$

we have, using (13),

$$\mu_2 + \mu_5 = \mu_6 + \mu_3$$

The non-zero stresses given by (33) are then of the form

$$\sigma_{11} = \sigma_{22} = \sigma_{33},$$
$$\sigma_{12} = \sigma_{21} = \mu\dot{\gamma} \tag{35}$$

where

$$\mu = \mu_4 + \mu_6 + \mu_3 = \mu_4 + \mu_5 + \mu_2$$

Thus the fluid behaves as a Newtonian fluid. If ϵ is non-zero, but small, there will be small differences in the normal stresses. We should then expect slight normal stress effects of the general type found in polymer solutions. As far as I know, normal stress measurements have not been made. With respect to independence of apparent viscosity on shear rate, at higher rates of shear, theory and experiment are in accord. Clearly, a measurement of the μ in (35) tells us little about the moduli involved in the theory.

As is discussed by Leslie,[8] the familiar correlations between Poiseuille flow, Couette flow and simple shear apply for the simplified theory here examined. In terms of the more general theory there are questions concerning Poiseuille flow. Here, the shear rate is not high near the center line, so use of the simplified theory is suspect. From Atkin's[15] work on this, **d** is parallel to the streamlines at the center line, whether ϵ be zero or not. According to the simplified theory, it is not, unless ϵ be zero. This raises some doubt as to the validity of concluding that $\epsilon = 0$ from an examination of capillary viscometer data alone. Porter and Johnson[12] note that consistent values of apparent viscosity obtain from capillary and concentric cylinder instruments, which lends credence to the notion that ϵ is small, if not zero.

Leslie[17] and Ericksen[18] have given analyses concerning the possible occurrence of instabilities in Couette flow, resembling the Taylor instabilities for Newtonian fluids, though the subject is still in a somewhat tentative state. One difficulty is that the analysis involves a narrow gap approximation, but does not account for the previously discussed complications associated with small gaps.

Effect of Magnetic Fields

With respect to exploring effects of static magnetic fields in viscometry, theory has lagged behind experiments such as are mentioned by Porter and Johnson.[12] Interpretation of experiments has involved the plausible assumption that a sufficiently strong field will align **d** parallel to it. An elementary theory of this is discussed by Ericksen.[19] For it, the estimates (17) and (19) are used to account for the effects of the field. Generally, if the field is in the plane of shear, **d** will be urged by the flow to take one orientation, by the field to take another and will compromise at an intermediate orientation. For a fixed liquid and fixed direction of the field, the orientation attained depends, theoretically, on the value of the ratio

$$R = H^2/\dot{\gamma}$$

H being the field strength. When the field is perpendicular to the plane of shear, there is no compromise, **d** being purely flow oriented or parallel to the field, depending on the value of R. As $R \to \infty$, **d** becomes parallel to the field, assuming appropriate values of susceptibilities. As $R \to 0$ it approaches the orientation obtaining when $H = 0$, discussed above. In the former limit, we know **d**, so there is the possibility of measuring various stresses to determine some of the μ's, as indicated in (33). Apparent viscosity measurements could give as many as four combinations. Likely possibilities are

$$\mathbf{d} = (0,0,1), \qquad 2\mu = \mu_4,$$
$$\mathbf{d} = (0,1,0), \qquad 2\mu = \mu_4 + \mu_5 - \mu_2,$$
$$\mathbf{d} = (1,0,0), \qquad 2\mu = \mu_4 + \mu_6 + \mu_3,$$
$$\mathbf{d} = 1/2(\sqrt{2}, \sqrt{2}, 0), \qquad 4\mu = \mu_1 + 2\mu_4 + \mu_5 - \mu_2 + \mu_6 + \mu_3$$

The last possibility might be of interest for normal stress measurements. In the first three cases, the normal stresses are all equal.

By a combination of apparent viscosity and normal stress measurements, at these four orientations, one might, in principle, determine all six μ's, though this has not been done.

A different check of the theory could be made by varying R, holding the direction of the field fixed. Apparent viscosity should then reduce to a function of R. Given data concerning moduli, this function could be calculated. Of course, we should avoid size effects such as were discussed earlier. As far as the magnetic field is concerned, static experiments and theory such as are discussed by Saupe[9] indicate that Hh, h being the gap width, should not be too small. Neither should $\dot{\gamma}h^2$. Since

$$R = H^2/\dot{\gamma} = (Hh)^2/\dot{\gamma}h^2$$

R is not restricted by these requirements.

An elementary theory for electric fields results from replacing the magnetic field intensity by the electric field, magnetization by polarization. From discussions such as are given by Brown and Shaw[20] (*cf.* Ch. VII), this theory is, if anything, less adequate than that for magnetism. Even so, there seems to be a range of conditions where apparent viscosity depends primarily on the corresponding ratio, the squared field strength divided by shear rate, according to the measurements of Björnstahl and Snellman.[21]

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